

Holographic image storage and multiple hologram storage in a planar Methyl Red-doped liquid crystal film

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Holographic images are recorded and the diffraction characteristic of the hologram is investigated in a planar Methyl Red-doped liquid crystal film without an external electric field. Multiple holograms are stored in this film with no external electric field, and peristrophic multiplexing, angular multiplexing, and spatial multiplexing are demonstrated to be suited for the implementation of azo-dye doped liquid crystal films among the multiplexing techniques. The diffraction efficiency is theoretically studied for multiple storage. Optical reconstruction for peristrophic multiplexed holograms and angular multiplexed holograms is discussed, which involves the peristrophic rotation angles and the recording angles, respectively. © 2008 Optical Society of America

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1. Introduction

Dye-doped nematic liquid crystals, which have many advantages over their inorganic counterparts and polymer-dispersed liquid crystals, such as high optical nonlinearity [1–4], low cost, ease of preparation and modification, and low external electric field, have attracted considerable attention [5–7,4,8–11]. Laser-induced dynamic gratings [5–7] and optical wave mixing in some of these nonlinear optical media have been investigated in the literature [10–13]. Recent studies have demonstrated extraordinarily large optical nonlinearity of nematic liquid crystals doped with dye molecules arising from laser-induced director axis reorientation [9,12,14]. Holographic illumination with a gratinglike variation of the light intensity leads to the formation of a transient grating or a permanent grating in these liquid crystal films [3,14–16]. Therefore, dye-doped liquid crystals are excellent materials for use in holographic and diffractive optical devices, among which Methyl Red (MR)-doped liquid crystal is a special medium. Under the

different experimental conditions, the transient grating or the permanent grating can be created in this film [1,15].

MR is an azo dye with both *trans* and *cis* isomers. Induced by a polarized light, azo groups undergo an isomerization process (*trans*–*cis*–*trans*) and stably lie in a new orientation with respect to the original orientation. Because azo-dye molecules interact with liquid crystal molecules, liquid crystal molecules also undergo reorientation. Generally, holographic gratings in a dye-doped liquid crystal film are generated based on the following two processes: surface memory of molecular alignment and azo-dye-driven liquid crystal reorientation. Surface memory, which provides the anchoring force for alignment, reflects the interaction of the liquid crystal molecules with dye molecules on the inner surface of the cell. Reorienting dye molecules result in a stable reorientation of the liquid crystal director [14]. Especially, in the planar MR-doped liquid crystal film, the surface grating, in combination with the refractive-index modulation generated in liquid crystal, can function as a permanent grating [15]. Therefore holographic optical storage with long-term stability [16] can be

performed in this film. Furthermore, multiplexing techniques have been proposed to increase the storage density, such as spatial multiplexing [17], angular multiplexing [18], wavelength multiplexing [19], and peristrophic multiplexing [20]. However, multiple image storage has not been studied in these films.

In this paper, we study holographic storage and the diffraction behavior of the hologram in the planar MR-doped liquid crystal film. We also achieve multiple hologram storage by peristrophic multiplexing, angular multiplexing, and spatial multiplexing in the film. The experimental result reveals that the image storage capacity reaches 10 Mbits/cm² in the thin film by angular multiplexing and spatial multiplexing or by peristrophic multiplexing and spatial multiplexing. The diffraction efficiency for multiple storage and optical reconstruction for peristrophic multiplexed holograms and angular multiplexed holograms are discussed.

2. Sample and Experimental Setup

The fabrication process of the sample is as follows. First, two glass substrates are coated with a polymer PVA (polyvinyl alcohol, Aldrich) and then rubbed with a lens tissue in the same direction. Second, both substrates with the same rubbing direction are stuck to each other, and a cell is formed. The thickness of the cell is controlled to be 36 μ m by mylar spacers. Third, the cell is filled by capillarity with a mixture of 5CB (Aldrich) doped with a small amount of MR (1% by weight, Aldrich). This process creates elongated stress and strain on the polymer and facilitates the alignment of the long axis of the liquid crystal molecules along the rubbed direction [21].

Figure 1 illustrates the experimental setup for holographic image storage. M_1 , M_2 , and M_3 are mirrors, BS is a beam splitter, $\lambda/2$'s are half-wave plates, SLF is a spatial light filter, and L is a lens with a focal length of 500 mm. IP and OP are the input and output planes, respectively. A reference beam (I_1) and a signal beam (I_2), which are derived from an Ar⁺ laser ($\lambda = 488$ nm) with exposure intensities and diameters 20 mW/cm², 2.5 mm and 18 mW/cm², 2 mm at the film, respectively, are set to be *s* polarized by half-wave plates and intersect in the sample. The sample with the horizontal director vector of liquid crystal is placed in the Fourier plane of the signal beam. And there is no electric field applied to the film. Input images are set at the input plane. A He-Ne laser beam

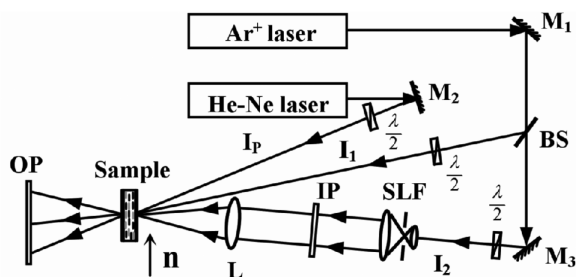


Fig. 1. Experimental setup for holographic image storage.

(I_p), which is also set to be *s* polarized with an intensity of 10 mW/cm² and a diameter of 2 mm, is used to probe the writing region. The angle between I_p and the bisector of the recording angle is 2.8°, and the incident plane is perpendicular to the surface of the sample. The output images are recorded on the screen at the output plane without a lens.

3. Holographic Image Storage

Usually, the reference beam is a plane wave in the holographic recording. When the signal beam is also a plane wave carrying no spatial information, the recorded grating in the film is a pure sinusoidal variation of the index of refraction in the extreme case. When the signal beam with spatial information of a general object illuminates the sample, a hologram is formed in the film [22]. The hologram, which must consist of several gratings, is recorded in the planar MR-doped liquid crystal film with a small recording angle of 4° without an external field applied, as shown in Fig. 2, which illustrates the procedures of holographic image storage and reconstruction.

Figures 2(a) and 2(b) show the images recorded by use of an Ar⁺ laser with the direction of the average long molecular axis of the liquid crystal, called the director *n*, parallel to the light polarization direction under illumination for only 12 minutes and 18 minutes, respectively, in which the second-order diffraction can be found. The second-order reconstructed images are about twice the size of the first-order images or the incident image, which can be used in holographic image amplification [23]. However, in the cases of too big incident images or too small recording angles, they usually overlap with the first-order diffraction images, as shown in Fig. 2(a). In fact, the distance between the *m*-order and the (*m* + 1)-order diffractions (*m* = 0, 1, 2, ...) increases with the increase of the recording angle. If necessary, the second-order reconstructed images can be spatially distinguished from the first-order ones by minimizing the incident image or increasing the recording angle, as shown in Fig. 2(b).

Figures 2(c) and 2(d) are the reconstructed images from the recorded holograms by a He-Ne laser with the diffraction efficiencies of 3.3% and 3.7%, respectively. Here, the diffraction efficiency is given by $\eta = I'/I_{\text{incident}} \times 100\%$ at the wavelength of the readout light, where I' is the intensity of the first-order diffraction beam, and I_{incident} is the incident intensity of the readout light. Experimental results show that high-order reconstructed images are observed with a strong recording intensity or a long enough exposure time. The +*m*-order and the -*m*-order diffraction images are centrosymmetrically inverted and therefore can be applied in holographic image rotation [23]. These two holograms have been stored for six months without obvious change.

4. High-Order Diffraction Image

As mentioned above, the high-order diffraction images are different from the low-order diffraction

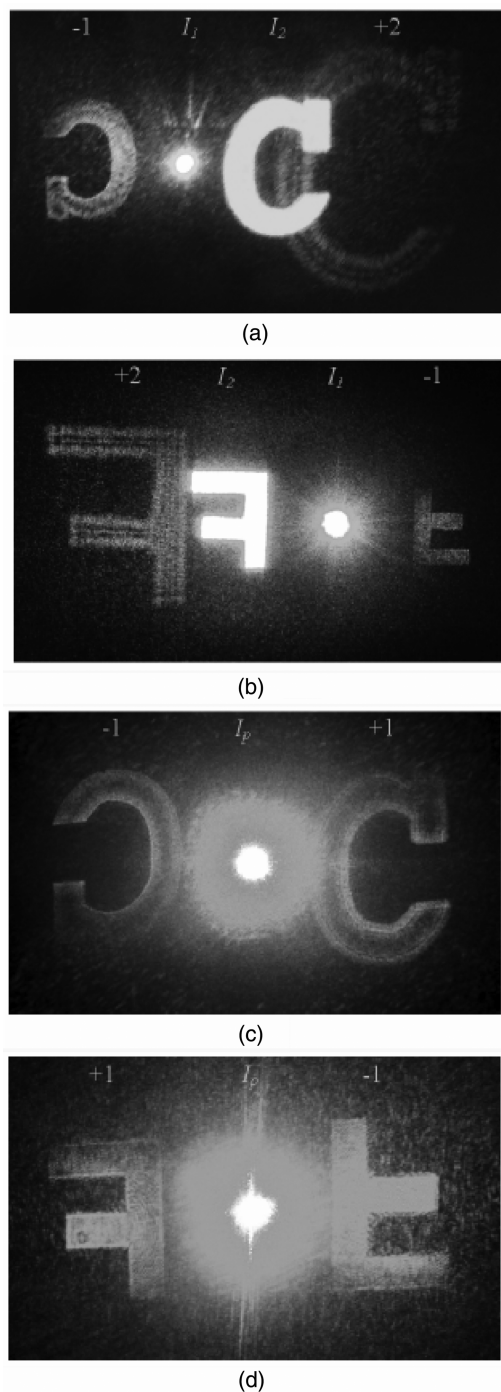


Fig. 2. Photographs of holographic image storage and reconstruction.

images in size. Here, the equations to calculate the size of each-order diffraction images are achieved. Figure 3 shows the sketch of higher-order diffracted images and the wave-vector diagram.

Our work has achieved that the diffraction angles depend on the recording angle for a recorded grating. The grating spacing is written as [21]

$$\Lambda = \frac{\lambda}{2n \sin \theta_0}, \quad (1)$$

where $2\theta_0$ is the included angle between the two recording beams. The parameter n is the refractive index of the film at the recording wavelength. The symbol λ is the wavelength of the recording light.

Then, the requirements for constructive interference of the recording light wave can be fulfilled only at particular angles, which is expressed as [22]

$$\begin{aligned} \Lambda \sin \theta_0 + \Lambda \sin \theta_m &= m \frac{\lambda}{n}, \\ m &= 1, 2, 3 \dots, \end{aligned} \quad (2)$$

where θ_m is the diffraction angle for a recording light, which is the angle between the m -order diffraction beam and the reference light.

Equation (1) is substituted into Eq. (2), and then the diffraction angle θ_m is determined by

$$\begin{aligned} \sin \theta_m &= (2m - 1) \sin \theta_0, \\ m &= 1, 2, 3 \dots. \end{aligned} \quad (3)$$

We assume that the wave vector of the reference beam is $\mathbf{k}_1$, and the wave vector of the focused signal beam ranges between $\mathbf{k}_2$ and $\mathbf{k}_3$. We define the intersecting angle between $\mathbf{k}_2$ and $\mathbf{k}_3$ as θ_2 and that between $\mathbf{k}_1$ and $\mathbf{k}_2$ as θ_1 . Then, the incident angle is given by $\theta'_0 = \theta_1/2$ for the wave coupling of $\mathbf{k}_1$ and $\mathbf{k}_2$ and $\theta''_0 = 1/2(\theta_1 + \theta_2)$ for the wave coupling of $\mathbf{k}_1$ and $\mathbf{k}_3$. L is the distance between the sample and the screen. In the case of a small recording angle, the m -order diffraction angle of $\mathbf{k}_1$ and $\mathbf{k}_2$ is $(2m - 1)\theta'_0$ and that of $\mathbf{k}_1$ and $\mathbf{k}_3$ is $(2m - 1)\theta''_0$. The parameter $\Delta\theta$ is the inclusion angle between the grating vectors $\mathbf{K}_1$ and $\mathbf{K}_2$, which are formed by $\mathbf{k}_1$ and $\mathbf{k}_2$, $\mathbf{k}_1$ and $\mathbf{k}_3$, respectively.

The size of the fundamental frequency image can be expressed as

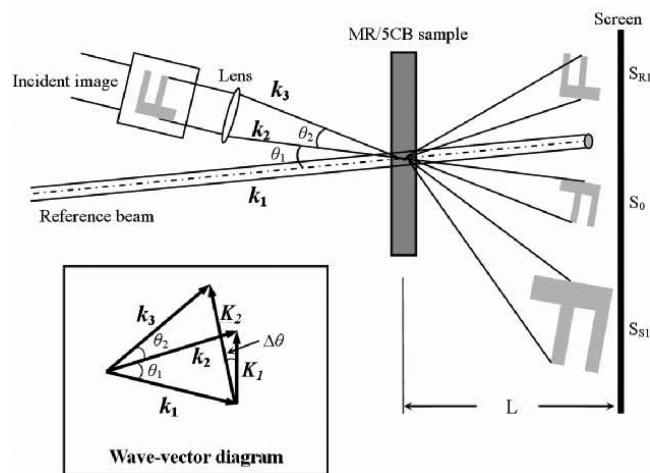


Fig. 3. Sketch of higher-order diffracted images and wave-vector diagram.

$$S_0 = L[\tan(\theta'_0 + \Delta\theta) - \tan(\theta'_0)] \\ = L\left[\tan\left(\theta_2 + \frac{\theta_1}{2}\right) - \tan\left(\frac{\theta_1}{2}\right)\right], \quad (4)$$

which can be rewritten as $S_0 \approx L\theta_1$ for the small recording angle.

The size of the m -order diffraction image at the side of the signal beam can be given by

$$S_{sm} = L\{\tan[(2m-1)\theta'_0 + \Delta\theta] - \tan[(2m-1)\theta'_0]\} \\ = L\left\{\tan\left[m\theta_2 + \left(m - \frac{1}{2}\right)\theta_1\right] - \tan\left[\left(m - \frac{1}{2}\right)\theta_1\right]\right\}, \\ m = 1, 2, 3, \dots \quad (5)$$

Under the condition of the small recording angle, it can be approximated as $S_{sm} \approx mL\theta_2$.

The size of a high-order reconstructed image at the side of the reference beam can be written as

$$S_{Rm} = L\{\tan[(2m-1)\theta'_0 - \Delta\theta] - \tan[(2m-1)\theta'_0]\} \\ = L\left\{\tan\left[(m-1)\theta_2 + \left(m - \frac{1}{2}\right)\theta_1\right] - \tan\left[\left(m - \frac{1}{2}\right)\theta_1\right]\right\}, \\ m = 1, 2, 3, \dots \quad (6)$$

This equation can be approximated to $S_{Rm} \approx (m-1)L\theta_2$ for the small recording angle.

Then, the ration of the size of the high-order diffraction image at the side of the signal light and the fundamental frequency image is $S_{Om}/S_0 \approx m$, which means that the size of the high-order reconstructed image is an integral multiple of the size of the fundamental frequency image. When the ratio is $S_{Rm}/S_0 \approx m-1$, the size of the high-order diffraction image is also an integral multiple of the size of the fundamental frequency image; and when $S_{Rm}/S_0 \approx 0$, the m -order diffraction image at the side of the reference beam is just the reference light.

The Eqs. (4)–(6) can be used to calculate the size of the high-order diffraction image for the case that the signal and reference beams intersect in the film at a small recording angle and at a focus plane of the Fourier lens. In the cases that the two beams intersect in the sample after and before the focus point of the Fourier lens, the offset of the focus point should be taken in. Usually, the reference beam is not an expanded light but a line beam laser, and the written region or the signal light spot in the sample is smaller than the reference light spot, which mean that the recording plane should be much closer to the focus plane of the lens. Then, the offset of the focus point is much smaller than the focal length of the Fourier lens so that the offset can be neglected.

5. Multiple Hologram Storage

Among the multiplexing techniques, peristrophic multiplexing is particularly suited for the implementation of the thin holographic recording medium,

which is achieved when the reference beam is rotated around the film's surface normal after each hologram is recorded or when the film is rotated around the reference beam. Angular multiplexing is the most commonly used scheme, and the recording beams for each page should be incident at the same position on the recording medium with different incident angles.

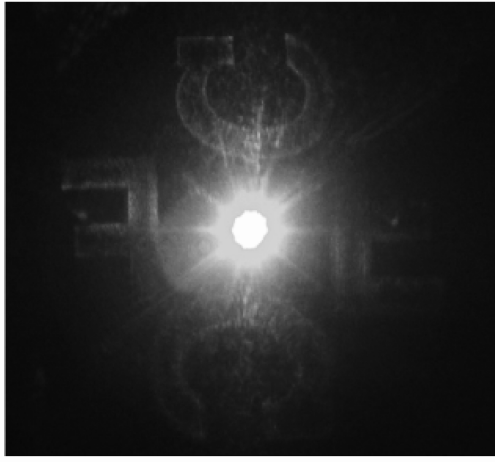
Here, two holograms have been stored at one location by peristrophic multiplexing. Figure 4 illustrates the photographs of multiple image reconstructions by an argon laser and by a He–Ne laser. After the letter “C” has been recorded for 20 minutes at the recording angle of 4° with the director n parallel to the light polarization direction, the film is rotated around the reference beam to its new position with the director n perpendicular to the light polarization direction, and then the letter “F” is stored for the same exposure time. As shown in Fig. 4, both images are read out with the director n perpendicular to the polarization direction. The average diffraction efficiencies of the +1st-order and the –1st-order diffractions of “C” and “F” are, respectively, 1.2% and 1.1% at 633 nm, which are much higher than those of 0.3% and 0.2% at 488 nm, respectively, so that the reconstructed images is usually read by He–Ne laser.

Also, two images have been recorded at a location by angular multiplexing with the horizontal liquid crystal director and both s -polarized writing beams. Multiple images reconstructed by He–Ne laser is shown in Fig. 5. The letter “B” and the heart shape have been written for 20 minutes at the recording angles, 4.8° and 2.2° , respectively, with their average diffraction efficiencies of the +1-order and the –1-order reconstructed images of 0.9% and 1.4% at 633 nm. In addition, these peristrophic-multiplexed or angular-multiplexed holograms can be stored for at least six months.

In this system, the film has a high degree of freedom of movement in rotational and linear movement. In order to increase the capacity, spatial multiplexing can be applied along with peristrophic multiplexing or angular multiplexing, in which multiple multiplexed holograms are stored at multiple locations. The total capacity of the holographic memory system is calculated by multiplying the capacity of peristrophic multiplexing or angular multiplexing by the numbers from the spatial multiplexing. The storage capacity C is given by [18]

$$C = NM, \quad (7)$$

where N is the number of bits in each stored page and M is the number of superimposed pages. Assuming one bit per pixel, N is limited to about 10^6 by current spatial light modulator technology. Since M is equal to 2 because of two recorded holograms at a location, the total storage capacity at one location with an area of 20 mm^2 is 2 Mbits. Then, the storage density reaches 10 Mbits/cm^2 . This work reveals that 50 pages of holographic images can be stored in our film with an



(a)



(b)

Fig. 4. Multiple image reconstructions (a) by argon laser and (b) by He-Ne laser.

area of 5 cm^2 by spatial and peristrophic multiplexings or by spatial and angular multiplexings.

MR-doped liquid crystals possess large birefringence, $n_{\parallel} - n_{\perp} = 0.2 \sim 0.3$, in the visible to infrared spectral regime, where $n_{\parallel}$ and $n_{\perp}$ are the refractive indices for the light polarization direction parallel and perpendicular to the director vector, respectively. The diffraction efficiency is given by $\eta \cong (\pi d \Delta n / \lambda)^2$, where d is the sample thickness, λ is the wavelength of the diffraction light, and Δn is the light-induced refractive index change [4,21].

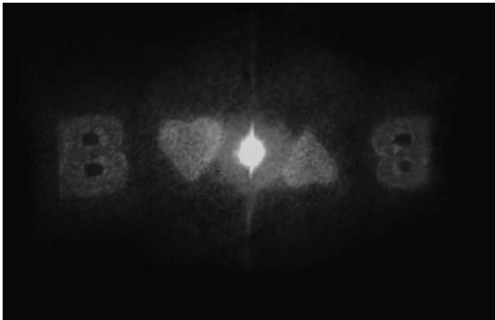


Fig. 5. Multiple images reconstructed by He-Ne laser.

When the other grating is stored at the location, the recording beams not only write the second grating but also erase the first grating. The diffraction efficiencies of both gratings are

$$\eta_1 \cong \left(\frac{\pi d}{\lambda} \Delta n_1 \right)^2 = \left[\frac{\pi d}{\lambda} \left(\Delta n - \Delta n^{\text{Erase}} \right) \right]^2, \quad (8)$$

$$\eta_2 \cong \left(\frac{\pi d}{\lambda} \Delta n_2 \right)^2. \quad (9)$$

In fact, the refractive index changes, Δn , Δn^{Erase} , and Δn_2 , depend not only on their inducing light intensities but also on dye concentration, incident angle, the angle between the polarization direction of the light and the director vector of the film, and recording time [3].

Assuming M is large enough, which is the number of gratings stored at a location, then the average diffraction efficiency is

$$\bar{\eta} \cong \left(\frac{\pi d}{\lambda} \Delta \bar{n} \right)^2, \quad (10)$$

where $\Delta \bar{n} = \Delta n_{\text{max}} / M$. Since the maximum refractive index change of 7.45×10^{-3} is obtained in MR-doped liquid crystal [1,15], theoretically, the number of holograms at a single location can be improved to 24 with an average diffraction efficiency of 0.3%, which is high enough to reconstruct a hologram, and the storage density can reach 0.12 Gbit/cm^2 . Here, the physical limitations of optical devices limit the storage capacity. When the recording angles cover all the optimum range of 0.5 to 5° with a long focal-length lens, and the diffraction efficiency is increased by a higher-power Ar^+ laser, the storage density will be greatly improved.

Due to the lack of Bragg selectivity, the reconstructed images from multiple holograms in the Raman-Nath regime appear simultaneously. We find two methods to solve the problems of optical reconstruction. Multiple-order diffractions from a hologram are in the incident plane of both writing beams. Peristrophic multiplexed holograms with different incident planes can be distinguished by peristrophic rotation angles. For angular multiplexed holograms with the same incident plane, the diffraction angles are different, and depend on their recording angles at a small probe angle.

The diffraction angle θ'_m , which is the angle between the m -order probe diffraction beam and the normal to the diffraction grating [24], is obtained by substituting the grating spacing, $\Lambda = \lambda / (2n \sin \theta)$, into the requirements for constructive interference of the probe light wave, $\Lambda \sin \theta_p + \Lambda \sin \theta'_m = m \lambda_p / n_p$.

$$\sin \theta'_m = 2mn\lambda_p \sin \theta / (n_p \lambda) - \sin \theta_p, \quad (11)$$

$$m = 0, 1, 2, \dots,$$

where θ and θ_p are the incident and probe angles, respectively, and n and n_p are the refractive indices of the film at the recording wavelength of λ and probe wavelength of λ_p , respectively. Since the incident angles are different, every-order reconstructed images from angular multiplexed holograms must be discriminated from the others with different diffraction angles.

6. Conclusions

We record holographic images with great readout image quality and study high-order diffraction images in planar MR-doped nematic liquid crystal film without an external electric field, and which have the potential applications in holographic image rotation and amplification. Also, we store multiple holograms with excellent optical stability in this film with no external electric field by peristrophic multiplexing, angular multiplexing, and spatial multiplexing. The diffraction efficiency is discussed for multiple storage. Also, two efficient and simple methods to solve the problems of optical reconstruction for angular and peristrophic multiplexed holograms are presented, which involve recording angles and peristrophic rotation angles, respectively. These theoretical results may provide a basis for an optical address recognition scheme of multiple holograms in azo-dye doped liquid crystals.

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